

<b>TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED / ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371</b>		ATTORNEY'S DOCKET NUMBER <b>P67314US0</b>
		US APPLICATION NO. (If known, see 37 CFR 1.5) <b>09/926683</b>
INTERNATIONAL APPLICATION NO. <b>PCT/EP00/03799</b> ✓	INTERNATIONAL FILING DATE <b>27 April 2000</b> ✓	PRIORITY DATE CLAIMED <b>1 June 1999</b> ✓
TITLE OF INVENTION <b>MIXTURE OF METAL AND/OR ALLOY PARTICLES AND A LIQUID MEDIUM, AND PROCESS FOR THE PREPARATION THEREOF</b>		
APPLICANT(S) FOR DO/EO/US <b>Wolfgang GLAESER</b> ✓		

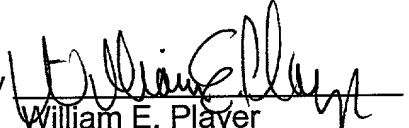
**Applicant herein submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information.**

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. ☒ A proper Demand for Internatl. Preliminary Examination was made by the 19th month from earliest claimed priority date.
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☒ has been transmitted by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
  - b. ☐ have been transmitted by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☒ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ A translation of the annexes to the Internatl. Preliminary Examination report under PCT Article 36 (35 U.S.C. 371(c)(5)).

**Items 11. to 16. below concern other document(s) or information included:**

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.  
☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☒ Other items or information:

International Search Report – EPO  
 PCT/IB/301 Form  
 PCT/IB/304 Form  
 PCT/IB/308 Form  
 First Page of Publication  
 International Preliminary Examination Report – No Annexes

US APPLICATION NO (If known, see 37 CFR 1.5) <b>09/926683</b>		INTERNATIONAL APPLICATION NO <b>PCT/EP00/03799</b>		ATTORNEY'S DOCKET NUMBER <b>P67314US0</b>	
17. <input checked="" type="checkbox"/> The following fees are submitted: <b>Basic National Fee (37 CFR 1.492(a)(1)-(5)):</b> Internatl. prelim. examination fee paid to USPTO (37 CFR 1.492 (a) (1)) .. \$710.00 No international preliminary examination fee paid to USPTO (37 CFR 1.492 (a) (2)) but international search fee paid to USPTO (37 CFR 1.445(a)(2)) .. \$740.00 Neither international preliminary examination fee (37 CFR 1.492 (a) (3)) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO) ..... <b>\$1040.00</b> International preliminary examination fee paid to USPTO (37 CFR 1.492 (a) (4)) and all claims satisfied provisions of PCT Article 33(2)-(4) ..... \$100.00 Search Report prepared by the EPO or JPO (37 CFR 1.492 (a) (5)) ..... <b>\$890.00</b> <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				CALCULATIONS	PTO USE ONLY
				\$ 890.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).				\$	
<b>Claims</b>	<b>Number Filed</b>	<b>Number Extra</b>	<b>Rate</b>		
Total Claims	7 - 20 =	-0-	x \$18.00	\$	
Independent Claims	1 - 3 =	-0-	x \$84.00	\$	
Multiple Dependent Claim(s) (if applicable)			+ \$280.00	\$	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				\$ 890.00	
Reduction by 1/2 for filing by <b>small entity</b> , if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				\$	
<b>SUBTOTAL =</b>				\$ 890.00	
Processing fee of \$130 for furnishing the <b>English translation</b> later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f))				\$	
<b>TOTAL NATIONAL FEE =</b>				\$ 890.00	
Fee of \$40.00 for recording the enclosed <b>assignment</b> (37 CFR 1.21(h)). Assignment must be accompanied by appropriate cover sheet (37 CFR 3.28, 3.31).				\$	
<b>TOTAL FEES ENCLOSED =</b>				\$ 890.00	
				Amt. to be refunded:	\$
				Amt. charged:	\$
a. <input checked="" type="checkbox"/> A check in the amount of \$ <u>890.00</u> to cover the above fees is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. <u>06-1358</u> in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge my account any additional fees set forth in §1.492 during the pendency of this application, or credit any overpayment to Deposit Account No. <u>06-1358</u> . A duplicate copy of this sheet is enclosed.					
SEND ALL CORRESPONDENCE TO:  <b>JACOBSON HOLMAN PLLC</b> 400 7th Street, N.W., Suite 600 Washington, DC 20004 202-638-6666 <b>CUSTOMER NUMBER: 00136</b>					
				By  William E. Player Reg. No. 31,409	

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Wolfgang GLAESER

Serial No.: New

Filing Date: December 3, 2001

For: MIXTURE OF METAL AND/OR ALLOY PARTICLES AND A  
LIQUID MEDIUM, AND PROCESS FOR THE  
PREPARATION THEREOF

PRELIMINARY AMENDMENT

Assistant Commissioner of Patents  
Washington, D.C. 20231

Sir:

Prior to initial examination, please amend the above-  
identified application as follows:

IN THE SPECIFICATION

Please insert the following sentence on line 1, immediately  
following the title:

--This is a 371 of PCT/EP00/03799, filed April 27, 2000,  
the disclosure of which is incorporated herein by reference.--

IN THE CLAIMS

Please amend claims 3-5 and 7 as follows:

## CLAIMS

3. (amended) The mixture according to claim 1, characterized in that said mixture exhibits a direct contact between almost all of the particles.
4. (amended) The mixture according to claim 1, characterized in that said electrolytic medium contains no or just a little gelling agent.
5. (amended) The mixture according to claim 1, characterized in that said metal and/or alloy particles consist of zinc or a zinc alloy.
7. (amended) A process for preparing the mixture according to claim 1, characterized in that a dosable mixture with excess amounts of the electrolytic medium is prepared first, which excess is sucked off from the mixture later after the dosing.

REMARKS

The foregoing Preliminary Amendment is requested in order to delete the multiple dependent claims and avoid paying the multiple dependent claims fee.

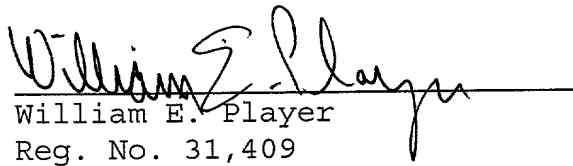
Attached hereto is a marked-up version of the changes made to the specification and claims by the current amendment. The attached page is captioned VERSION WITH MARKINGS TO SHOW CHANGES MADE.

Early action on the merits is respectfully requested.

Respectfully submitted,

JACOBSON HOLMAN PLLC

By

  
William E. Player  
Reg. No. 31,409

400 Seventh Street, N.W.  
Washington, D.C. 20004-2201  
(202) 638-6666

Atty. Docket: P67314US0  
Date: December 3, 2001  
WEP:jrc

VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE CLAIMS

3. (amended) The mixture according to claim 1 ~~or 2~~, characterized in that said mixture exhibits a direct contact between almost all of the particles.
4. (amended) The mixture according to claim 1 ~~any of claims 1 to 3~~, characterized in that said electrolytic medium contains no or just a little gelling agent.
5. (amended) The mixture according to claim 1 ~~any of claims 1 to 4~~, characterized in that said metal and/or alloy particles consist of zinc or a zinc alloy.
7. (amended) A process for preparing the mixture according to claim 1 ~~any of claims 1 to 6~~, characterized in that a dosable mixture with excess amounts of the electrolytic medium is prepared first, which excess is sucked off from the mixture later after the dosing.

SMB

Mixture of Metal and/or Alloy Particles and a Liquid Medium,  
and Process for the Preparation Thereof

The present invention relates to a mixture of metal and/or alloy particles and a liquid electrolytic medium, and a process for the preparation thereof. Such mixtures are known and are used for different purposes, those mixtures being particularly important in which the metal and/or alloy particles can dissolve in the liquid electrolytic medium and thereby release electric current. Thus, in particular, these are mixtures of metal or alloy particles in which the particles can dissolve in an acid or lye. Such chemical reactions are extensively used for the production of electric current, for example, in batteries and accumulators, i.e., rechargeable batteries.

A very frequently employed mixture consists of zinc particles or particles of a zinc alloy and solutions of alkali as the liquid electrolytic medium. These mixtures are employed, for example, together with mixtures of manganese dioxide with components of a liquid electrolytic medium, the two mixtures mostly being separated by a separator. The particles of zinc or zinc alloys are prepared from liquid zinc or liquid zinc alloys according to various known methods, for example, by atomization or granulation on a rotating granulating dish. Depending on the process conditions, the grain sizes, the grain size distribution and the outer shapes of the particles can be adjusted, sieve fractions often being separated from the oversize and undersize.

From WO 99/07030, it is known to selectively admix more or less superfine grains with the per se known particles of zinc or zinc alloys, because this is supposed to reduce the sensitivity towards mechanical shocks. It is believed that these properties can be attributed to the fact that a more favorable ratio of volume to surface

of the particles is provided and the properties of the batteries are thereby improved, and/or the superfine grain provides for better contacts between the particles.

For stabilizing the mixtures of particulate zinc and an electric electrolyte, various methods have been known. Thus, for example, DE-PS-818 519 describes the addition of paper fibers and a gelling agent for dry cell electrodes. DE-AS-25 10 934 describes a water-insoluble cross-linked polyacrylamide as a binder, indicating that the previously employed binders, such as sodium carboxymethylcellulose, did not meet with the desired success.

It has been the object of the invention to provide mixtures of metal and/or alloy particles and a liquid electrolytic medium which, for example, when incorporated in batteries and accumulators, exhibit optimum properties in terms of performance, durability, resistance to heavy discharges and mechanical shocks while the release of gases is avoided.

This object has now been achieved by the volume of the liquid electrolytic medium approximately corresponding to the spaces between the particles in a dry packing (for example, according to ASTM-B 212). This is equivalent to the demand that the volume of the mixture be approximately equal to or little larger than the volume of a dry packing of the metal and/or alloy particles. In these mixtures, it is practically ensured that there is a direct contact between almost all particles while there is still enough liquid electrolytic medium to dissolve the metal and/or alloy particles to produce current. In contrast, according to the prior art, the mixture contains from about 50 to 100% more liquid electrolyte than would fit into the spaces between the particles in a dry packing. Accordingly, the volume of such mixtures is significantly higher than would correspond to the volume of the particles in a dry packing.

A possible drawback of the mixtures according to the invention is the fact that they cannot easily be dosed in the usual way due to their being neither flowable nor free-flowing, but having a relatively solid consistency. Therefore, to be able to prepare these mixtures and fill them in a dosed manner, for example, into batter-



ies, it is possible to first prepare a mixture with excess amounts of the electrolytic medium and to take care that this excess can flow from the mixture later after the dosing or is sucked off. When batteries based on particles of zinc or zinc alloys and manganese dioxide are prepared, this can be achieved relatively easily by partly or completely dispensing with moistening the separator, for example, made of paper or non-woven materials, after the manganese dioxide has been introduced and the separator inserted, and the amount of liquid medium to be employed is first used for rendering the mixture of metal and/or alloy particles and the liquid electrolytic medium more easily dosable.

Then, after this dosable mixture has been introduced, the excess amount of the electrolytic medium is sucked off through the separator and also supplied to the manganese dioxide. Thus, the finished battery contains amounts of electrolytic medium which are altogether comparable with those used for batteries prepared by previous methods. In contrast, the mixture of metal and/or alloy particles no longer contains the usual volume excess of liquid electrolytic medium, which has previously been chosen for the mixture to be readily dosable. However, the volume of the liquid medium did not fit into the free pore volume of the particles in a dry packing.

To prevent the later settling of the metal and/or alloy particles in the electrolytic media, a gelling agent has previously been added to these mixtures. However, both the gelling agent and the excess amount of electrolytic medium can affect the properties of the finished battery and even deteriorate certain properties. For example, not all of the particles are in immediate contact with other particles, so that there is no electronic conductivity between them. Thus, the internal resistance of the battery increases. The addition of gelling agent can reduce the electric conductivity of the electrolytic medium. Further, all previously prepared batteries have been more or less sensitive towards heavy mechanical impacts and shocks.

The use of the mixtures according to the invention, in which the volume of the medium approximately corresponds to the spaces between the particles in a dry packing, provides a practically direct metallic contact between almost all particles.

This reduces the internal resistance of the battery and, above all, also reduces the sensitivity towards shocks.

Since the aggregation of these particles is relatively stable, the use of gelling agent can even be dispensed with more or less. At any rate, the amount of gelling agent added can be significantly reduced. However, the previously usual amounts may also be added, if desired.

A dry packing is a still relatively loose aggregation. By intensively vibrating and shaking, such dry packings can altogether be further condensed. However, since a sufficient amount of liquid electrolytic medium is also necessary in batteries, the maximum possible condensation of the metal and/or alloy particles is avoided. However, the previously employed significant volume excess of liquid electrolytic medium is also avoided. According to the invention, the mixtures are first rendered dosable with excess amounts of the electrolytic medium, then dosed and thereafter freed from the excess amounts of electrolytic medium. In practice, this is effected by suction, wherein this superfluous amount is taken up by the separator and cathode, for example.

The bulk density of a dry packing, for example, according to ASTM-B 212, is highly dependent on the grain size, the grain size distribution and the outer shape of the particles. In order to achieve, on the one hand, the advantage of the relatively stable structure with metallic contact between the particles, and on the other hand, to be able to accommodate sufficient amounts of liquid electrolytic medium in the mixture, it is recommendable, when zinc or zinc alloys are used, to employ a material which has a dry bulk density of lower than 2.8 g/ml. This is possible, for example, when a material is used which contains many elongated or elongated-flat particles due to its production method, because this results in a lower bulk density. This means that there is a relatively large pore volume between the particles. When such materials are used, it is altogether possible then to prepare a comparable weight distribution between the metal particles and the liquid electrolytic medium in the finished battery, as in the previously prepared batteries. This distribution of weights has proven useful in principle, and therefore the battery manufacturers would not much like to substantially change it. According to the

invention, the manner of dosing the mixtures of metal and/or alloy particles and the liquid electrolytic medium needs not to be changed either. However, the moistening of the separator with impregnation electrolyte may be changed. This step may either be completely omitted, or performed with a clearly lower amount of impregnation electrolyte, because the mixture of metal and/or alloy particles and the liquid electrolytic medium as used according to the invention is capable of releasing the excess electrolytic medium to the separator and cathode. In addition, the preparation of the batteries in the various standardized sizes can still be effected in an unchanged manner. For example, the mercury-free zinc alloys usual today, which are little gassing and are environment-friendly, can be employed. However, the mixtures of metal and/or alloy particles and a liquid electrolytic medium according to the invention may also be employed in other types of batteries and accumulators to bring about novel and advantageous properties of the batteries and accumulators.

CLAIMS:

1. A mixture of metal and/or alloy particles and a liquid electrolytic medium, characterized in that the volume of the medium approximately corresponds to the spaces between the particles in a dry packing.
2. The mixture according to claim 1, characterized in that the volume of the mixture is approximately equal to or little larger than the volume of a dry packing of the metal and/or alloy particles.
3. The mixture according to claim 1 or 2, characterized in that said mixture exhibits a direct contact between almost all of the particles.
4. The mixture according to any of claims 1 to 3, characterized in that said electrolytic medium contains no or just a little gelling agent.
5. The mixture according to any of claims 1 to 4, characterized in that said metal and/or alloy particles consist of zinc or a zinc alloy.
6. The mixture according to claim 5, characterized in that the particles have a bulk density of lower than 2.8 g/ml.
7. A process for preparing the mixture according to any of claims 1 to 6, characterized in that a dosable mixture with excess amounts of the electrolytic medium is prepared first, which excess is sucked off from the mixture later after the dosing.

07262645

**DECLARATION  
AND POWER OF ATTORNEY  
U.S.A.**

FOR ATTORNEYS' USE ONLY

ATTORNEYS' DOCKET NO.

ALL PATENTS, INCLUDING DESIGN

FOR APPLICATION BASED ON PCT; PARIS CONVENTION;

NON PRIORITY; OR PROVISIONAL APPLICATIONS

As a below named inventor, I declare that my residence, post office address and citizenship are stated below next to my name, the information given herein is true, that I believe that I am the original, first and sole inventor (if only one name is listed at 201 below), or an original, first and joint inventor (if plural inventors are named below at 201-203, or on additional sheets attached hereto) of the subject matter which is claimed and for which patent is sought on the invention entitled:

Mixture of metal and/or alloy particles and a liquid medium, and process for the preparation thereof

which is described and claimed in: ☒ PCT International Application No. PCT/EP 00/03799 filed 27/04/2000  
☐ the attached specification ☐ the specification in application Serial No. \_\_\_\_\_ filed \_\_\_\_\_  
(if applicable) and amended on \_\_\_\_\_

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 (a)-(d) of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)  
199 25 048.8 Germany 01/06/1999  
(Number) (Country) (Day/Month/Year Filed)  
☒ Yes ☐ No

(Number) (Country) (Day/Month/Year Filed)  
☐ Yes ☐ No

(Number) (Country) (Day/Month/Year Filed)  
☐ Yes ☐ No

I hereby claim the benefit under Title 35, United States Code, §119(e) of any United States provisional application(s) listed below:

Application No. \_\_\_\_\_ Filing Date \_\_\_\_\_ Application No. \_\_\_\_\_ Filing Date \_\_\_\_\_

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)

(Filing Date)

(Status: patented, pending, abandoned)

8 POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorneys (Registration No.) to prosecute this application, receive and act on instructions from my agent, and transact all business in the Patent and Trademark Office connected therewith. HARVEY B. JACOBSON, JR. (20,851); D. DOUGLAS PRICE (24,514); JOHN CLARKE HOLMAN (22,769); MARVIN R. STERN (20,640); MICHAEL R. SLOBASKY (26,421); JONATHAN L. SCHERER (29, 851); IRWIN M. AISENBERG (19,007); WILLIAM E. PLAYER (31,409)

SEND CORRESPONDENCE TO:

**JACOBSON, PRICE, HOLMAN & STERN**  
PROFESSIONAL LIMITED LIABILITY COMPANY  
400 Seventh Street, N.W.  
Washington, D.C. 20004

DIRECT TELEPHONE CALLS TO:

(please use Attorney's Docket No.) (202) 638-6666

**JACOBSON, PRICE, HOLMAN & STERN**  
PROFESSIONAL LIMITED LIABILITY COMPANY

1-00 Inventor(s) name must include at least one unabbreviated first or middle name.

201	FULL NAME* OF INVENTOR	FAMILY NAME <u>GLAESER</u>	GIVEN NAME <u>Wolfgang</u>	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY <u>Goslar</u> <u>DEX</u>	STATE OR FOREIGN COUNTRY <u>Germany</u>	COUNTRY OF CITIZENSHIP <u>Germany</u> ✓
	POST OFFICE ADDRESS	POST OFFICE ADDRESS <u>Sieben Linden 7</u>	CITY <u>Goslar</u>	STATE OR COUNTRY <u>Germany</u> ZIP CODE <u>38640</u>
202	FULL NAME* OF INVENTOR	FAMILY NAME	GIVEN NAME	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY	STATE OR COUNTRY ZIP CODE
203	FULL NAME* OF INVENTOR	FAMILY NAME	GIVEN NAME	MIDDLE NAME
	RESIDENCE & CITIZENSHIP	CITY	STATE OR FOREIGN COUNTRY	COUNTRY OF CITIZENSHIP
	POST OFFICE ADDRESS	POST OFFICE ADDRESS	CITY	STATE OR COUNTRY ZIP CODE

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under section 1001 of Title 18 of the United States Code; and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon

SIGNATURE OF INVENTOR 201*	SIGNATURE OF INVENTOR 202*	SIGNATURE OF INVENTOR 203*
<u>[Signature]</u>		
DATE <u>27.10.01</u>	DATE	DATE

☐ Additional inventors are named on separately numbered sheets attached hereto.

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